1 Response to Anonymous Referee #1. We thank the anonymous reviewer for an insightful and thoughtful 2 review. Below are our responses.

3

4 Referee 1: Are fluctuations in calculated CO2 caused by layer-specific differences in stomatal data 5 or delta13C, or both? Are there significant differences in stomatal conductance or delta13C?

6

7 Author response: Indeed, in the traditional approach to atmospheric CO₂ reconstructions using changes in 8 plant physiology, inferred CO₂ variations can be traced directly to either leaf carbon isotopic composition 9 or changes in stomatal density. However, the approach that we try to advocate relies on gas-exchange 10 modeling and it 1) is sensitive to any combination of changes in carbon isotopic composition and stomatal conductance, and 2) takes into account the cumulative response of different plant species (i.e. all the plant 11 12 species determined from the plant fossil locality). This approach thereby accounts for the complexities 13 that arise from non-linear, and even non-uniform physiological responses to changes in the climate, something that CO₂ reconstructions using only stomata, or only leaf δ^{13} C values, and only a single species 14 cannot do. Moreover, as we note in the manuscript this comprehensive approach leads to a more accurate 15

accounting of uncertainty in ultimate CO₂ estimates than traditional approaches. 16

17

18 However, to address the reviewer's question, we conducted ANOVA linked with TukevHSD to test

differences in leaf δ^{13} C and Stomatal Density between zones. We approached the ANOVA – TukeyHSD 19 20 with three different null hypotheses (H₀): 1) leaf δ^{13} C and stomatal density combining all species is the

same for all zones, 2) leaf δ^{13} C and stomatal density for all canopy species, after Z-score scaling of inter-21

species variation, is the same for all zones, and 3) leaf δ^{13} C and stomatal density for the most abundant 22

23 species, Litsea calicarioides, is the same for all zones. The p-value in all cases is higher than 0.05, indicating that H₀ cannot be rejected in any of these scenarios, and that leaf δ^{13} C and stomatal density do not 24

25 individually change significantly between zones. Thus, variations in estimated CO₂ are the result of the

26 combination of leaf carbon isotopic composition, leaf conductance, and intra-species variation of

27 physiological response to atmospheric carbon. The original carbon isotope and leaf conductance

28 measurements are available in the supplementary material. We include new a section in the manuscript to

further clarify how our approach means that a change in model output may be impossible to trace to a 29

- uniform change in input variables, and on a related note we emphasized the need for further evidence to 30 31 further evaluate the role of a CO₂ increase in driving Antarctic Ice melt at the Oligocene/Miocene
- 32 boundary.
- 33

34 Referee 1: It would be also interesting to compare stomatal data of the fossil plants with those of 35 their extant representatives. Are there significant differences?

36

37 Author response: We agree that this is an interesting research question, and it is currently considered in 38 the context of a separate study. The comparison between fossil plants and their extant representatives is 39 not of fundamental relevance to this manuscript and we prefer to keep it separate from the research results 40 we are reporting here.

41

42 Referee 1: The treatment of intrinsic Water-Use Efficiency is too simplistic and should include 43 consideration of the changes in fatty acid δD of the Foulden Maar record, in particular with

44 regards to the influence of changes in humidity on plant water-use efficiency reconstructions.

45

Author response: We do have δD values and $\delta^{13}C$ values from leaf waxes in this record that can provide 46

some guidance for making inferences about changes in hydroclimate across the 100,000-vr period of 47

sedimentation (Reichgelt et al., 2016). However, our discussion of iWUE is not meant to address 48

49 variations that occurred *during* this interval, but instead focuses on contrasting the early Miocene values

with modern values. To support our southern temperate reconstructed iWUE, we include results from the 50

same transform functions on previously published records from Ethiopia and Panama, which showed 51

52 53 54 55 56 57 58	similar offsets from modern. That said, we agree with the referee that in a warmer world, whether you are in the tropics or in the southern temperate region, you would expect higher vapor pressure deficits, which would also drive up the iWUE signal. We have therefore expanded the discussion to address this uncertainty and included Fig. S3 in the supplement to show that while temperatures Miocene New Zealand are higher than modern, the relative humidity reconstructed for Foulden Maar is well within the range of modern New Zealand forested biomes.
59 60 61 62 63	Referee 1: It is difficult to extrapolate leaf-level productivity to the canopy and vegetation level. It is suggested that the authors mention and discuss the research on modern CO2 fertilization experiments that highlight the complexity of physiological response in forests to increased atmospheric carbon dioxide.
64 65 66	Author response: We expanded discussion on the confounding factors observed in modern CO2 fertilization experiments.
67 68 69	Referee 1: P. 2, l. 42 "will make more C available to the terrestrial biosphere". This is an awkward description of the anticipated fertilization effect of elevated CO2.
70 71	Author response: This sentence has been amended for clarity.
72	Referee 1: P. 4, l. 98 "For conductance measurements" This is not exactly correct. With fossil
73 74	leaves, anatomical data are determined which then allow to approximate conductance (on the basis of various assumptions). This is not the same as measuring conductance of living leaves P 4 1 103
75 76	See previous comment.
77 78	Author response: amended.
79 80	Referee 1: P. 8, ls. 194 – 195 There seems to be something wrong with the structure of this sentence.
81 82	Author response: amended.
83 84 85 86	Referee 1: P. 10, l. 229 - 231 "including a measure for the relative time the leaf is assimilating". What is the final value for this relative time? How was it determined? Additionally, the symbol for this relative time appears to be the same as for the operational conductance.
87 88	Author response: amended.
89 90 91 92 93	Referee 1: P. 10, ls. 238 - 239 "is derived from Maire et al. (2015) which included coordinates, habit, An and Gw data from which we could then calculate" It is not clear (from this sentence), how the calculations were conducted in detail. Why were "coordinates" used and for what? Why where Gw data from Maire et al. (and therefore of extant plants) used, and not conductance data derived from stomatal data of the fossil plants?
94 95 96	Author response: amended.
97 98 99 100 101	Referee 1: P. 15, ls. 355 – 357 "In contrast to iWUE Gw for Miocene trees is similar to the modern day range." Since Gw is derived from Gc and therefore from fossil material, this would mean that "structural" conductance is not that different for the fossil plants and their extant relatives?

102 103 104 105 106	Author response: That is correct. We have expanded the discussion on this. We note (here and in the manuscript) that the extant relatives are not the same as the plants that currently occur at this latitude. Due to cooling the warm-temperate to subtropical diverse Lauraceae dominated rainforests of Miocene New Zealand no longer exist.
107 108 109 110	Referee 1: P. 15, ls. 357 - 359 "Increased atmospheric evaporative demand in combination with a longer growing season". The authors describe that they used CLAMP to reconstruct growing season length. As far as I know, CLAMP provides also data on humidity. See also general comments
111	comments.
112	Author response: Thanks for this excellent suggestion CLAMP data on humidity have now been included
112 113 114	in the supplementary material and are now included in our discussion.
115	
116	
117	
118 119	Response to Referee #2, Gregory Retallack. Many thanks to Dr. Retallack for his insightful review. His comments are greatly appreciated.
120	
121	Reviewer comment: This paper is well written and relatively free of errors, and presumably has
122	been reviewed before.
123	
124	Author response: It had not been reviewed before and we appreciate compliment.
125	
126	Reviewer comment: Earth System Sensitivity (1.242) is a very slippery concept in this context,
127	because the temperature increase with CO2 doubling in any one part of the world will depend on
128	where it is. There are already numerous studies showing that midlatitude continental sites show
129	little temperature change and thus muted sensitivity, but tropical and polar sites show marked
130	changes in temperature. New Zealand is a temperate, site but also globally unusual in having a
131	strongly marine-influenced climate now, and even more so in the Miocene when there was little
132	and and few mountains. It is not clear now this even becomes relevant later (1.5/4) where elevated
133	CO2 estimated is thought to relate to ESS of 4-/oC, because Miocene paleotemperature for New
134	Zealand is not ollered.
126	Author response: Perhaps this wasn't clear enough in our discussion. We agree that local temperatures are
130	not particularly instructive when considering ESS. The temperature change we considered was an
138	estimate of the global average based on data and the transfer function approach presented in Hansen et al.
139	(2013) We have updated the text to clarify this While this approach itself has its limitations (which we
140	also discuss in the manuscript), we believe that it is a reasonable basis for the ESS discussion and that this
141	discussion is valuable.
142	
143	Reviewer comment: I fail to see the relevance of C4 grassland expansion (l. 388) because it
144	postdated the age of these New Zealand leaves by some 10 million years.
145	
146	Author response: We updated the text to better explain the relevance of C4 grassland expansion in the late
147	Miocene in the context of our results.
148	
149	Reviewer comment: Errors in estimated paleoatmospheric CO2 are asymmetric and very large (l.
150	307-8). Perhaps this is due to inadequate numbers of stomates counted: it should be hundreds in
151 152	each image. Furthermore, Gaussian error propagation can be used to calculate symmetrical errors. Both issues are addressed in the following paper too recent to be included - Retallack, G.J. and

Conde, G.D., 2020. Deep time perspective on rising atmospheric CO2. Global and Planetary Change, p.103177.

154 155

156 Author response: The reviewer raises an interesting point about the numbers of stomata per image

- 157 counted resulting in larger error bars. We have now added text to further clarify how we avoided
- systematic error propagation and we now have referenced the reviewer's suggested study to highlight the
- 159 importance of the number of counts per image.
- 160 The asymmetrical error propagation is the result of the gas-exchange model iteratively solving for 161 conductance, assimilation rate and carbon dioxide, centered around a representative assimilation rate (A_0)
- 162 measured under modern day atmospheric carbon dioxide. Because of this approach, the model returns a
- 163 greater number of divergent solutions for fossil conductance, assimilation rate, and carbon dioxide for 164 high CO₂ estimates, and fewer for low CO₂ estimates.
- 165
- 166
- 167
- 168

- 169 Elevated CO₂, increased leaf-level productivity and water-use efficiency during the early Miocene
 170
- 171 Tammo Reichgelt^{1,2}, William J. D'Andrea¹, Ailín del C. Valdivia-McCarthy¹, Bethany R.S. Fox³, Jennifer
- 172 M. Bannister⁴, John G. Conran⁵, William G. Lee^{6,7}, Daphne E. Lee⁸
- 173
- ¹Lamont-Doherty Earth Observatory, Columbia University, Palisades, New York, USA.
- ²Department of Geosciences, University of Connecticut, Storrs, Connecticut, USA.
- ³Department of Biological and Geographical Sciences, University of Huddersfield, Huddersfield, UK.
- ⁴Department of Botany, University of Otago, Dunedin, New Zealand.
- 178 ⁵ACEBB & SGC, School of Biological Sciences, The University of Adelaide, Adelaide, Australia.
- ⁶Landcare Research, Dunedin, New Zealand.
- ⁷School of Biological Sciences, University of Auckland, Auckland, New Zealand.
- ⁸Department of Geology, University of Otago, Dunedin, New Zealand.
- 182 Correspondence: Tammo Reichgelt (tammo.reichgelt@uconn.edu)
- 183
- 184 Abstract. Rising atmospheric CO₂ is expected to increase global temperatures, plant water-use efficiency, 185 and carbon storage in the terrestrial biosphere. A CO_2 fertilization effect on terrestrial vegetation is 186 predicted to cause global greening as the potential ecospace for forests expands. However, leaf-level fertilization effects, such as increased productivity and water-use efficiency, have not been documented 187 from fossil leaves in periods of heightened atmospheric CO_2 . Here, we use leaf gas-exchange modeling 188 on a well-preserved fossil flora from early Miocene New Zealand, as well as two previously published 189 190 tropical floras from the same time period, to reconstruct atmospheric CO₂, leaf-level productivity, and intrinsic water-use efficiency. Leaf gas-exchange rates reconstructed from early Miocene fossils which 191 192 grew at southern temperate and tropical latitudes, when global average temperatures were $5-6^{\circ}$ C higher 193 than today reveal that atmospheric CO₂ was ~450–550 ppm. Early Miocene CO₂ was similar to projected values for 2040AD, and is consistent with Earth System Sensitivity of $3-7^{\circ}$ C to a doubling of CO₂. The 194

195 Southern Hemisphere temperate leaves had higher reconstructed productivity than modern analogs likely 196 due to a longer growing season. This higher productivity was presumably mirrored at northern temperate 197 latitudes as well, where a greater availability of landmass would have led to increased carbon storage in forest biomass relative to today. Intrinsic water-use efficiency of both temperate and tropical forest trees 198 199 was high, toward the upper limit of the range for modern trees, which likely expanded the habitable range 200 in regions that could not support forests with high moisture demands under lower atmospheric CO₂. 201 Overall, early Miocene elevated atmospheric CO₂ sustained globally higher temperatures and our results 202 provide the first empirical evidence of concomitant enhanced intrinsic water-use efficiency, indicating a 203 forest fertilization effect.

204

205 1 Introduction

206 Terrestrial plants comprise 450 Gt of carbon, representing 80% of Earth's dry carbon (C) biomass (Bar-on 207 et al., 2018). Globally, plants draw down \sim 120 Gt of atmospheric C per year through photosynthesis, representing the largest annual C flux on Earth (Beer et al., 2010). Total plant biomass is believed to be 208 209 determined in large part by atmospheric carbon dioxide concentrations (C_a), and it is predicted that future 210 increases in C_a will have a three-pronged effect on the terrestrial biosphere: 1) increased global 211 temperatures will shift the boundaries of climate zones and thereby the potential forest expanse (Rubel 212 and Kottek, 2010); 2) productivity will increase because global photosynthesis is C limited and increased C_a will have a fertilization effect on the terrestrial biosphere (Zhu et al., 2016); and 3) elevated C_a will 213 214 increase plant water-use efficiency and reduce the threshold for physiological drought (Cernusak, 2020), 215 making more land area available for biosphere expansion (Zhou et al., 2017). Plant fossils record the effect of past changes in climate, including CO₂ enrichment, and thus fossil floras provide insight into 216 217 changes in the carbon cycle and their effects on the terrestrial biosphere from a natural, whole-ecosystem 218 perspective.

The Miocene has been considered problematic for our understanding of Earth System Sensitivity
(ESS) to C_a, because most proxy-based C_a estimates indicate concentrations near 300 ppm (Foster et al.,

221 2017), close to pre-industrial values, yet global temperatures were 5–6 °C higher than modern (Hansen et 222 al., 2013). Enhanced radiative forcing is required to maintain such elevated early Miocene temperatures 223 (Herold et al., 2010; Hansen et al., 2013), and without elevated C_a, climate models cannot achieve such 224 high global temperatures in the Miocene (Henrot et al., 2010). The early Miocene also had an expanded 225 biosphere compared to today, including woody vegetation in locations that are currently too cold and/or 226 too dry for forests (e.g. Askin and Raine, 2000; Herold et al., 2010). A biosphere of the magnitude 227 observed in the early Miocene fossil record requires elevated temperatures and plant water-use efficiency, 228 suggesting C_a levels higher than estimated by most existing proxy reconstructions (Herold et al., 2010; Henrot et al., 2010). Importantly, plant-based C_a reconstructions have challenged the consensus of low C_a 229 in the early Miocene (Kürschner and Kvaček, 2009; Reichgelt et al., 2016; Tesfamichael et al., 2017; 230 Londoño et al., 2018) and previous interpretations of alkenone-based C_a proxies are being disputed 231 232 (Bolton et al., 2016; Witkowski et al., 2019). 233 We applied plant gas-exchange modeling (Franks et al., 2014) to fossil leaves from the early Miocene

(~23 Ma) rainforest ecosystem from southern New Zealand preserved in the Foulden Maar deposit 234 (Bannister et al., 2012; Reichgelt et al., 2013; Conran et al., 2014; Lee et al., 2016) to reconstruct carbon 235 236 assimilation rates (A_n), intrinsic water-use efficiency (iWUE; the ratio between carbon assimilation and 237 stomatal conductance to water), and the C_a levels required to maintain these values. The same analyses were performed on previously published leaf δ^{13} C and micromorphological measurements from two early 238 Miocene fossil floras from Ethiopia (Tesfamichael et al., 2017) and Panama (Londoño et al., 2018). These 239 two studies were incorporated here because they represent a similar time period (early Miocene) and the 240 241 authors applied the same gas-exchange methodology. Because A_n , iWUE, and C_a are interdependent (Farquhar et al., 1980; Drake et al., 1997), we reconstructed these variables in concert for each fossilized 242 leaf recovered from these forest ecosystems. This allows us to make inferences concerning carbon 243 244 availability, productivity, and water balance in the forest.

247 2.1 Site Description

248Foulden Maar (Fig. 1a) is a unique Konservat-Lagerstätte with abundantly preserved plants and insects

249 (Kaulfuss et al., 2015; Lee et al., 2016) in southern New Zealand (45.527°S, 170.219°E). It was formed in

- an ancient maar-diatreme lake (Fig. 1b) at the Oligocene/Miocene boundary (Fox et al., 2015; Kaulfuss,
- 251 2017) and consists of ~100 kyr of annually laminated diatomite (Lindqvist and Lee, 2009; Fox et al.,
- 252 2016). The Foulden maar-diatreme complex is part of the larger late Oligocene late Miocene Waipiata
- 253 Volcanic Field that produced a variety of maar volcanoes and scoria cones (Németh and White, 2003).
- Plant fossils used in this study were collected from a ~183 m long drill core (Fig. 1c). The Lauraceae-
- dominated rainforest (Bannister et al., 2012) surrounding the lake grew at a paleolatitude of ~50°S (Fig.
- 1a). The climate was marginally subtropical with a mean annual temperature of ~18°C, similar to modern

day climates at 30°S (Reichgelt et al., 2019). The length of the growing season in this climate was ~ 10

258 months, compared to 5–6 months today, as reconstructed from the surface exposure macrofossil

assemblage using the Climate Leaf Analysis Multivariate Program (Reichgelt et al., 2013).

260

261 **2.2** Fossil leaf anatomy and paleoecology

Mummified fossil leaves were extracted from turbidite deposits that occur frequently within the Foulden 262 263 Maar diatomite core (Fox et al., 2015). The diatomite is loosely consolidated allowing mummified leaves to be extracted using a combination of water and scalpels. After extraction, the leaves were cut into three 264 pieces: one for bulk δ^{13} C analysis, one for stomatal conductance measurements, and a third as a reference 265 specimen. Leaf δ^{13} C was measured using a Costech elemental combustion system (EA) coupled to a Delta 266 V Plus IRMS (Thermo). In order to place measured δ^{13} C on the VPDB scale, we calibrated measurements 267 using a two-point isotope calibration based on the USGS40 and USGS41 standards. Measurement 268 uncertainty was calculated by replicating ~ 15 samples two or three times and applying averaged 269 270 uncertainty to the remaining leaves. For stomatal density and geometry measurements the leaves were 271 soaked in hydrogen peroxide (H_2O_2) with up to 40% dilution, with tetra-sodium pyrophosphate salt crystals (Na₄P₂O₇ \cdot 10[H₂O]), on a boiling plate at 40–50 °C for 1–2 hours (Bannister et al., 2012). When 272

273 the adaxial and abaxial cuticle layer could be separated, the leaf layers were cleaned of mesophyll cell 274 debris using small paintbrushes and both layers were stained with <0.5% Crystal Violet ($C_{25}N_3H_{30}Cl$) and 275 mounted on glass slides with glycerin jelly. Stomatal conductance and geometry measurements were 276 made on pictures at 100× magnification using TSView 7.1.1.2 microscope imaging software on a Nikon 277 Optiphot. Leaves were often fragmented and the cuticle wrinkled because the leaves were deposited in 278 turbidites. Moreover, there was strong divergence in overall cell and stomatal density, because we made 279 measurements on all species recovered from the sediments. To avoid systematic errors arising from 280 wrinkled cuticle, differing leaf architecture between species, or low cell counts (Retallack and Conde, 281 2020), each picture was given a standard bounding box (0.3×0.3 mm) on which cells were counted, to calculate stomatal density. The number of cells in each bounding box ranged from 100 - 750, strongly 282 dependent on species. Five to eight pictures were taken of each leaf to constrain errors in cell density. 283 284 Stomatal size measurements were made using ImageJ 1.48v software (Schneider et al., 2012). 285 18 distinct leaf morphotypes were identified from the Foulden Maar drill core. Descriptions and justification for identification are found in the Supplementary Information. Species identifications are 286 287 provided, where possible, based on paleobotanical studies from the Foulden Maar surface exposures. 288 Known species recovered from the Foulden Maar drill core are *Litsea calicarioides* (Fig. S1a), 289 Cryptocarya taieriensis (Fig. S1b), C. maarensis (Fig. S1i), Beilschmiedia otagoensis (Fig. S2a) 290 (Lauraceae) (Bannister et al., 2012), Laurelia otagoensis (Fig. S2h) (Atherospermataceae) (Conran et al., 291 2013), and Hedycarva pluvisilva (Fig. S2i) (Monimiaceae) (Conran et al., 2016). Otherwise, tentative 292 genus or family identifications are provided, or unspecified morphotypic qualifiers, for leaves that could not be assigned a plant group. These will henceforth be referred to as "C" (Fig. S1c), cf. Myrtaceae (Fig. 293 S1d), cf. Ripogonum (Fig. S1e), cf. Myrsine (Fig. S1f), "H" (Fig. S1g), cf. Elaeocarpaceae/Cunoniaceae 294 (Fig. S1h), cf. Dysoxylum (Fig. S2b), cf. Cryptocarva (Fig. S2c), "O" (Fig. S2d), "P" (Fig. S2e), "Q" 295 296 (Fig. S2f) and cf. Endiandra (Fig. S2g). 297 We made 375 anatomical and 80 carbon isotope measurements on 72 organically preserved fossil

leaves representing the 18 species collected from the Foulden Maar deposit (Fig. 1a). The affinities of

299 modern living relatives of the plant types at Foulden Maar strongly suggest that during the Miocene the 300 site was characterized by a multi-layered closed canopy rainforest ecosystem (Reichgelt et al., 2013; 301 Conran et al., 2014). In order to determine atmospheric carbon (C_a), intrinsic water-use efficiency (iWUE), and carbon assimilation rates (A_n), the ecological strategies of the individual fossil species at 302 303 Foulden Maar need to first be established (Reichgelt and D'Andrea, 2019). Understory species rarely 304 experience light saturation and utilize respired CO_2 that has already undergone isotopic fractionation; 305 both conditions influence gas-exchange modelling results (Royer et al., 2019). Therefore, Ca reconstructed from understory species cannot be considered indicative of true global C_a. Here, we 306 determine whether a fossil leaf type was likely in the canopy or the understory, based on 1) leaf δ^{13} C, 2) 307 leaf cell density, and 3) sinuosity of the epidermal cell walls. A large range of leaf δ^{13} C in a single species 308 309 is indicative of different levels of light saturation, which indicates that this species may preferentially 310 occur in the subcanopy or in the understory (Graham et al., 2014). Leaves in the canopy, experiencing 311 light saturation, divide epidermal cells rapidly compared to leaves in the shade, leading to high cell 312 densities and relatively high leaf mass per areas in sun-exposed leaves (Šantrůček et al., 2014). Finally, a 313 high level of anticlinal cell wall sinuosity has been interpreted as indicative of low-light conditions 314 (Kürschner, 1997; Bush et al., 2017). We consider these three lines of evidence occurring in concert as 315 indicative of a canopy or subcanopy ecological preference.

316

317 2.3 Modelling gas-exchange

Atmospheric carbon dioxide (C_a), plant photosynthesis (A_n), and intrinsic water-use efficiency (iWUE) are tightly linked (Farquhar et al., 1980; Drake et al., 1997), which allows us to solve for these parameters iteratively, through anatomical and carbon isotope (δ^{13} C) measurements of the fossil leaves. The Franks et al. (2014) gas-exchange model solves for C_a , by iteratively reconstructing A_n and leaf conductance to atmospheric carbon (G_c), using a Monte Carlo approach. This means that every C_a reconstruction has an associated A_n and G_c value.

325
$$C_a = \frac{A_n}{G_c} \times \left(1 - \frac{C_i}{C_a}\right)$$
(1)

In which C_i/C_a represents the ratio of intercellular carbon to atmospheric carbon, which can be
reconstructed using known leaf fractionation processes: fractionation caused by diffusion (a),
carboxylation (b), and fractionation caused by the preferential uptake of ¹²C to ¹³C in photosynthesis (Δ),
which is also influenced by the rate at which the leaf is photosynthesizing (Farquhar et al., 1982).

$$\frac{C_i}{C_a} = \frac{\Delta - a}{b - a} (2)$$

333

Here, a = 4.4‰ and b = 29‰ (Farquhar et al., 1982; Roeske and O'Leary, 1984). Δ can be calculated from the δ^{13} C of the air, derived from Tipple et al. (2010) and measurements of leaf δ^{13} C (Farquhar and Richards, 1984; Farquhar et al., 1989). Leaf and air δ^{13} C used in the Franks et al. (2014) model are presented in Table S1.

338

339
$$\Delta = \frac{\delta^{13} C_{air} - \delta^{13} C_{leaf}}{1 + \delta^{13} C_{leaf}} (3)$$

340

G_c is determined by the maximum capacity for conductance of a leaf surface (G_{max}), the ratio of
operational conductance to G_{max} (ζ), boundary layer conductance (G_b), and mesophyll conductance (G_m)
(Franks et al., 2014).

344

345
$$G_c = \left(\frac{1}{G_b} + \frac{1}{\zeta \times G_{max}} + \frac{1}{G_m}\right)^{-1} (4)$$

G_b, ζ , and G_m are all changeable under natural conditions (e.g. Schuepp, 1993; Niinemets et al., 2009; 347 348 Londoño et al., 2018) and it is highly disputed if these variables can be determined from fossil leaf material at all (e.g. McElwain et a., 2016; Soh et al., 2017). However, we adopt a standardized approach 349 put forward by Franks et al. (2014) to obtain input for these variables. $G_b = 2 \pm 0.1 \text{ mol m}^{-2} \text{ s}^{-1}$, $\zeta = 0.2 \pm 0.1 \text{ mol m}^{-2} \text{ s}^{-1}$ 350 0.02 (Franks et al., 2009; Dow et al., 2014), and G_m is determined using an empirical calibration (Evans 351 and Von Caemmerer, 1996). 352 353 $G_m = 0.013 \times A_n$ (5) 354 355 G_{max} is determined using predominantly measurable anatomical features of the fossil leaf cuticle (Franks 356 and Beerling, 2009): stomatal density (SD), maximum aperture surface area (a_{max}), pore depth (p_d), and 357 the ratio of diffusivity of CO_2 in air over the molar volume of air (d/v), here taken as 0.000714 mol m⁻¹ s⁻¹ 358 359 (Marrero and Mason, 1972). 360 $G_{max} = d/_{\mathcal{V}} \times SD \times \frac{a_{max}}{p_d + \frac{\pi}{2}\sqrt{a_{max}/\pi}}$ (6) 361 362 In this equation, SD can be measured directly from the leaf, p_d is assumed to be the same as guard cell 363 364 width (gcw), and a_{max} is determined assuming a circular opening for the aperture, with the measurable pore length (p₁) as the diameter (Franks et al., 2014). 365 366 $a_{max} = \pi \times \frac{p_l}{4} \ (7)$ 367 368

Measurements of SD, gcw and p₁ used in the Franks et al. (2014) gas-exchange model are presented in
Table S1.

Because A_n is required to solve G_m , G_c is solved iteratively, though G_c is largely determined by measurable anatomical parameters. However, A_n is also solved iteratively, as it is dependent on C_a and the carbon saturation value (Γ), set at 40 ppm (Franks et al., 2013).

375

376
$$A_n \approx A_0 \times \frac{(C_a - \Gamma) \times (C_{a0} + 2\Gamma)}{(C_a + 2\Gamma) \times (C_{a0} - \Gamma)}$$
(8)

377

In which A_0 is the photosynthetic rate of a modern model species that can represent the fossil species' 378 379 photosynthetic rate, and Ca0 is the atmospheric carbon dioxide level at which A0 was measured. A0 for 380 each fossil species was derived from the compilation of photosynthetic rates presented in Reichgelt and 381 D'Andrea (2019). For fossil leaves with known modern relatives, we constrained the possible A_0 range by only including modern relatives within the same family or order, i.e. Lauraceae for Litsea calicarioides, 382 383 Cryptocarva taieriensis, C. maarensis, cf. Cryptocarva, Beilschmiedia otagoensis and cf. Endiandra, Myrtaceae for cf. Myrtaceae, Liliales for cf. Ripogonum, Primulaceae for cf. Myrsine, Elaeocarpaceae and 384 Cunoniaceae for cf. Elaeocarpaceae/Cunoniaceae, Meliaceae for cf. Meliaceae, Atherospermataceae for 385 Laurelia otagoensis, and Laurales for Hedycarya pluvisilva. Then, following the method of constraining 386 A_0 of modern living relatives presented in Reichgelt and D'Andrea (2019), only A_0 values of plants with 387 similar growth forms to the fossil plants, and growing in similar light environments as Foulden Maar were 388 389 included. A0 and Ca0 used in the Franks et al. (2014) model, and associated ecology of fossil leaf types is 390 shown in Table S2.

391

392 The Franks et al. (2014) gas-exchange model thus iteratively solves for C_a, A_n, and G_c. However, only

leaves derived from canopy trees are likely to represent these values at light saturation. Moreover, plants

- in the understory assimilate a mix of atmospheric and respired CO₂, which has therefore already
- 395 undergone fractionation processes, making the calculated C_i/C_a problematic. Therefore, we present the

396 results for Ca, An, and Gc of leaf types most likely to be derived from canopy trees separately, as they are 397 more likely to not have a systematic skew. iWUE is defined as the ratio between A_n and stomatal conductance to water (Feng, 1999). 398 399 $iWUE = \frac{A_n}{G_w} (9)$ 400 401 Due to the different rates at which carbon dioxide and water vapor diffuse in air, a transformation of G_c is 402 403 required to calculate G_w. 404 $G_w = 1.6 \times G_c (10)$ 405 406 Finally, cumulative annual carbon uptake through photosynthesis (A_{tot}) can be calculated in gC m⁻² yr⁻¹, 407 408 by transferring from moles to grams, including a measure for the relative time the leaf is assimilating 409 carbon (ζ), and the length of the growing season. 410 $A_{tot} = (2.6 \times \zeta \times A_n \times t_a) \times 12 (11)$ 411 412 413 In which t_g is the length of the growing season in months, which we can derive from the fossil plant 414 assemblage (Reichgelt et al., 2019), using the method of Spicer et al. (2009). Gw, Atot, and iWUE values 415 for Litsea calicarioides, Cryptocarya taieriensis, C. maarensis, cf. Elaeocarpaceae/Cunoniaceae, and cf. Myrtaceae are presented in Table S3. The modern reference A_n and G_w data are derived from Maire et al. 416 417 (2015), which, using transform functions 9 and 11, we also used to calculate iWUE and Atot. 418 2.4 Comparison to Earth System Sensitivity 419

atmospheric CO₂. This sensitivity is likely not static in Earth's history and is dependent on, among other
aspects, continental configuration and ocean circulation patterns (Royer, 2016). Here, we estimate global
surface temperature for the early Miocene following the approach of Hansen et al. (2013). We then use
these temperature estimates along with the a broad range of commonly cited Neogene ESS, of 3–7°C
(Hansen et al., 2013; Royer, 2016), to provide a model for the expected early Miocene C_a. Following this

Earth System Sensitivity to C_a (ESS) is the amount of temperature increase expected under a doubling of

 $\label{eq:compared} \mbox{model, ultimately means that a doubling of C_a compared to pre-industrial levels is expected when an$

427 increase of global average surface temperatures (T_s) of $3-7^{\circ}C$ compared to modern occurs.

428 Compiled deep-sea benthic foraminifera δ^{18} O data of the last 30 million years (Zachos et al., 2001) 429 were averaged into 20 kyr time bins. Deep-sea temperatures (T_d) were then calculated using the linear 430 transfer functions of Hansen et al. (2013), which depend on the presence of sea-ice.

431

420

432
$$T_d = 5 - 8 \times \frac{\delta^{18} 0 - 1.75}{3} IF \, (\delta^{18} 0 < 3.25)(12)$$

433
$$T_d = 1 - 4.4 \times \frac{\delta^{18} 0 - 3.25}{3} \, IF \, (\delta^{18} 0 > 3.25)(13)$$

434

435 T_s was then calculated for post-Pliocene using:

436

437
$$T_s = 2 \times T_d + 12.25$$
 (14)

438 For the Pliocene:

439	$T_s = 2.5 \times T_d + 12.15$ (1)	5)

- 440
- 441 And for pre-Pliocene we assumed that T_s changed linearly with T_d , by a factor of 1.5.
- 442

443
$$\Delta T_s = 1.5 \times \Delta T_d \ (16)$$

445 C_a based on an ESS range of 3–7°C was then calculated using the resulting T_s.

446

447
$$C_a = 310 \times \frac{T_{s[x]} - T_{s[0]}}{2 \times ESS} + 310 \ (17)$$

448

In which $T_{s[x]}$ is the calculated average global surface temperature at time x, $T_{s[0]}$ is the modern day average global surface temperature, and 310 represents pre-industrial C_a.

451

452 **3 Results and Discussion**

453 3.1 Southern Temperate Rainforest Paleoecology

454 Modern day Lauraceae rainforests in New Zealand have a single dominant canopy tree, Beilschmiedia

455 *tawa*, and its farthest southern extent is ~42°S (Leathwick, 2001), which is the farthest southern

456 occurrence of any arborescent Lauraceae species in the world. Rainforests further south in New Zealand

457 are usually dominated by Nothofagaceae or Podocarpaceae, and the only modern-day forests at ~50°S are

458 the Magellanic Subpolar Forests in southern South America. Low-growing Podocarpaceae/Nothofagaceae

459 forests, similar to modern forests in southern New Zealand and southern South America, dominated

460 Antarctic vegetation during the early Miocene (Askin and Raine, 2000) and the Foulden Maar rainforest

461 included at least ten Lauraceae species (Bannister et al., 2012), emphasizing the expanded biosphere

462 potential in the early Miocene compared to today (Herold et al., 2010).

463 We identify *Litsea calicarioides*, *Cryptocarya maarensis*, *C. taieriensis*, cf.

464 Elaeocarpaceae/Cunoniaceae, and cf. Myrtaceae as the most probable canopy components because they

lack characteristics typical of understory components, 1) the large range of leaf δ^{13} C values and relatively

- low overall leaf δ^{13} C values (Graham et al., 2014), 2) low cell densities (Kürschner, 1997; Bush et al.,
- 467 2017) (Fig. 2 a,b), and 3) the undulating or sinuous cell walls (Kürschner, 1997; Bush et al., 2017).
- 468 Modern day *Litsea calicaris* in New Zealand is also part of the canopy, though rarely dominant (de

469 Lange, 2020), whereas Cryptocarva is extinct in New Zealand. Members of Elaeocarpaceae, Cunoniaceae 470 and Myrtaceae in modern day New Zealand, such as Weinmannia racemosa (Cunoniaceae) and 471 Metrosideros robusta (Myrtaceae) can attain heights of over 25 meters (de Lange, 2020). The most likely subcanopy or understory taxa were cf. Ripogonum, cf. Myrsine, "O", and cf. 472 *Dysoxylum*, because leaf fossils of these types have both low leaf δ^{13} C and sinuous or undulating cells 473 474 (Fig. S1e,f, S2b,d). Ripogonum scandens in modern day New Zealand is a twining forest liana, often found in the understory, Myrsine comprises several species of shrubs and small trees; whereas Dysoxylum 475 476 spectabile in modern day New Zealand is a medium-sized tree (de Lange, 2020). The affinity of morphotype "O" is unclear, but likely represents a now extinct plant group in New Zealand. Hedycarya. 477 pluvisilva, Laurelia. otagoensis, cf. Cryptocarva, cf. Endiandra, Beilschmiedia. otagoensis, "C", "H", 478 "P", and "Q", all displayed some variation in these features and occurred in relatively low abundance, and 479 480 are therefore considered of uncertain ecological affinity.

481

482 **3.2 Earliest Miocene CO₂**

Gas-exchange modeling (Franks et al., 2014) of canopy leaves throughout the Foulden Maar core 483 484 indicates that C_a (±1 σ) was 445 +618 / -100 ppm, whereas reconstructed C_a from understory elements yields C_a of 622 +3017 / -161 ppm (Fig. 2c), consistent with understory plants assimilating respired CO₂ 485 that has undergone prior fractionation processes, as well as experiencing elevated levels of Ca under the 486 canopy (Graham et al., 2014; Royer et al., 2017). Prior work on the Foulden Maar core established three 487 different phases based on bulk organic δ^{13} C (Fig. 1c), fatty acid δ^{13} C, and fatty acid δ D: Phase I (80–105 488 m depth) with high δ^{13} C and low δ D, Phase II (55–65 m depth) with low δ^{13} C and high δ D, and Phase III 489 (0–45 m depth) with high δ^{13} C and low δ D (Reichgelt et al., 2016). Phase III can be further subdivided 490 into Phase IIIa (30–45 m depth) and IIIb (0–20 m depth), as Phase IIIa exhibits a period of low fatty acid 491 δ^{13} C and high δ D, which is not expressed in bulk organic δ^{13} C (Reichgelt et al., 2016). Gas-exchange 492 modelling on leaves from these phases (Fig. 1c) suggest that during Phase II and IIIa Ca may have been 493 elevated ($C_a = 529 + 1159 / -125$ and $C_a = 538 + 769 / -181$ ppm, respectively) compared to Phase I and 494

Phase IIIb ($C_a = 444 + 572 / -95$ and 442 + 1219 / -110 ppm, respectively) (Fig. 3). Although gas-exchange 495 modeling input reconstructed differing Ca between phases, differences in overall conductance parameters, 496 such as stomatal density and leaf δ^{13} C, are not apparent (Table S1), despite differences in bulk δ^{13} C, fatty 497 acid δ^{13} C, and δ D (Reichgelt et al., 2016). This is likely the result of non-uniform species responses to 498 499 environmental changes in a complex multi-layered rainforest ecosystem, such as at Foulden Maar. 500 The advantage of using gas-exchange modeling to reconstruct C_a from multiple species is that the 501 uncertainty is quantified and constrained, greatly reducing the potential for systematic error in the final 502 estimate (Reichgelt and D'Andrea, 2019; Royer et al., 2019). Along with the enhanced accuracy comes a 503 more comprehensive appraisal of uncertainty than is achieved using other proxy approaches (Fig. 4). 504 Proxy error propagation is based on mechanistic variability, grounded in known physical and physiological limits of plant gas-exchange that are understood to be universal (Franks et al., 2014). This 505 506 differs from empirical proxies, whose uncertainty representation is based on calibration error of modern-507 day observations without mechanistic constraints. Our canopy C_a estimate (445 +618 / -100 ppm, Fig. 2c) is independent of calibration error, based on universal gas-exchange mechanisms, and represents plant 508 509 vegetative organs of multiple plant species that directly interacted with the available pool of atmospheric 510 carbon dioxide. Previous C_a estimates from the Oligocene/Miocene boundary based on boron isotopes 511 and paleosol carbonates are generally lower than our estimates (Ji et al., 2018; Greenop et al., 2019) (Fig. 4b), whereas C_a estimates based on stomatal index and recent alkenone-based C_a estimates are more 512 similar to our results (Kürschner et al., 2008; Super et al., 2018). 513 Reconstructions of globally elevated temperatures of 5–6 °C in the early Miocene (Hansen et al., 514 2013) with a C_a of ~300 ppm (Ji et al., 2018; Greenop et al., 2019) upsets the expected ESS to C_a during 515 this period (Henrot et al., 2010). Geochemical Ca proxy estimates consistently produce Ca estimates that 516 are too low to satisfy ESS to C_a prior to the Pliocene (Royer, 2016) (Fig. 4a,b). Estimates from the fossil 517 518 leaf-based stomatal index proxy for C_a (Kürschner et al., 2008) on the other hand do indicate a positive 519 correlation between temperature and C_a in the Neogene (Fig. 4a). At present, there are too few studies that reconstruct C_a using gas-exchange modeling to allow for a full comparison to other C_a proxies; however, 520

521	our C_a estimates of ~450–550 ppm are in line with the ESS to C_a in the early Miocene (Fig. 4a,b), based
522	on modelling experiments (Herold et al., 2010; Henrot et al., 2010). Moreover, thus far, Neogene Ca
523	estimates reconstructed using gas-exchange methods (Reichgelt et al., 2016; Tesfamichael et al., 2017;
524	Londoño et al., 2018; Moraweck et al., 2019) appear to agree with the suggested ESS to C _a (Fig. 4a,b).
525	Bulk organic and leaf wax δ^{13} C values reveal a ~4‰ decrease at Foulden Maar over a 10-meter
526	interval at the beginning of Phase II (55–65 m depth), likely representing a time period of <10 kyr (Fox et
527	al., 2016). This shift in isotopic composition suggests a substantial change in the global carbon cycle
528	(Reichgelt et al., 2016). The mode of reconstructed values in this study suggests and increase of ~450 to
529	550 from Phase I to Phase II (Fig. 3). The C _a values stay near 550 ppm throughout Phase II and Phase
530	IIIa, representing a 20-40 kyr time period (Fig. 3). Absolute dating of Foulden Maar based on
531	paleomagnetic reversals in the core, annual lamination of lake sediments, and basalt-derived Ar/Ar dates
532	indicates that the deposition of the Foulden Maar sediment coincided with the termination of the earliest
533	Miocene (Mi-1) glaciation of Antarctica (Fox et al., 2015). Interestingly, an increase in C _a from ~450 to
534	\sim 550 ppm at the termination of Mi-1 is consistent with modeling studies indicating that C _a > 500 ppm is
535	necessary to terminate a large-scale Antarctic glaciation (DeConto et al., 2008). We note that with the
536	current data available, it is not possible to exclude the possibility that modeled Ca changes in the record
537	were influenced by canopy density changes or regional hydroclimate. However, our observations from
538	Foulden Maar are inconsistent with hydrological, ecological or Ca changes as the sole driver of plant
539	physiological response (Reichgelt et al., 2016), and it is more likely that two or more of these parameters
540	changed in concert.

542 **3.3 Elevated CO₂ and the early Miocene biosphere**

543 The Foulden Maar Miocene rainforest was primarily evergreen (Lee et al., 2016). The main Miocene

544 canopy trees at Foulden Maar, *Litsea calicarioides*, *Cryptocarya taieriensis*, *C. maarensis*, cf.

545 Elaeocarpaceae/Cunoniaceae and cf. Myrtaceae, had relatively high iWUE (Miocene iWUE first quartile

546 $[Q_1]$ - third quartile $[Q_3] = 70-101$) compared to modern evergreen trees (evergreen iWUE $Q_1-Q_3 = 31-100$)

547 73) (Fig. 5a). Reconstructed iWUE from tropical early Miocene plants (Tesfamichael et al., 2017; 548 Londoño et al., 2018) is slightly higher ($Q_1-Q_3 = 80-125$) (Fig. 5a). The difference between reconstructed Miocene iWUE and that of modern deciduous trees is greater still (deciduous iWUE $Q_1-Q_3 = 27-52$), 549 consistent with the expectation that increased C_a favors evergreen trees (Niinemets et al., 2011; Soh et al., 550 2019). In the method of reconstruction used here, iWUE is ultimately an expression of leaf δ^{13} C and 551 conductance (see Methods section 2.3). Therefore, similar to reconstructed Ca iWUE may be sensitive to 552 environmental factors other than C_a. For example, leaf δ^{13} C can change in response to edaphic conditions 553 and precipitation (e.g. Kohn, 2016; Cornwell et al., 2018), as well as vapor pressure deficit (VPD) (Franks 554 555 et al., 2013). The climate of Foulden Maar in the early Miocene was warm-temperate to subtropical, compared to the cool-temperate forests in southern New Zealand today (Reichgelt et al., 2019). Though 556 557 reconstructed relative humidity at Foulden Maar is within the range of modern New Zealand forest 558 biomes, the average monthly VPD was 500-700 Pa, compared to 250-450 Pa today (Fig. S3), which may 559 result in a similar reconstructed iWUE as under elevated C_a (Franks et al., 2013). However, the reconstructed ecosystem at Foulden Maar is a broad-leaved humid rainforest (Bannister et al., 2012), 560 which likely had a high annual moisture surplus (Reichgelt et al., 2019). Increased water-use efficiency in 561 response to relatively high VPD compared to modern would only be a positive trade-off if water 562 563 availability were limiting. Additionally, reconstructed iWUE from both temperate and tropical early Miocene floras are high compared to modern, suggesting a global signal, such as would be expected to 564 globally elevated C_a; not from VPD as the early Miocene tropics would not be warmer than today (Herold 565 566 et al., 2010). Most importantly, the modern iWUE data (Fig. 5a) are from a global database that includes 567 environments with annual moisture deficits (Maire et al., 2015). Because reconstructed early Miocene iWUE is higher even than modern plants experiencing high VPD, we argue that VPD differences cannot 568 explain the high iWUE values of the early Miocene, and that increased efficiency due to higher C_a is the 569 570 best explanation.

In contrast to iWUE, reconstructed conductance to water (G_w) for Miocene trees is similar to the
modern-day range at the same latitude (Fig. 5b), a somewhat surprising result because G_w is expected to

573 be reduced in high C_a climates (Franks and Beerling, 2009). Studies on modern forests also suggest the 574 absence of a reduction in Gw to enhanced Ca (Yang et al., 2016; Gimeno et al., 2018), or even an increase in the G_w (Frank et al., 2015). A longer growing season together with increasing VPD was proposed to 575 explain increasing G_w in modern European forests (Frank et al., 2015). Similarly, a relatively high water 576 577 flux from the forest to the atmosphere due to high water supply (Reichgelt et al., 2019) and high VPD (Fig. S3) could explain the broad similarity in the range of modern and early Miocene G_w, despite higher 578 Ca. The early Miocene Gw from tropical latitudes are within the range of modern evergreen tropical trees, 579 though relatively low (Early Miocene Q1-Q3: 0.08-0.13 mol m⁻² s⁻¹, modern evergreen Q1-Q3: 0.07-0.2 580 mol $m^{-2} s^{-1}$) (Fig. 5b). 581

582 A longer growing season likely resulted in the high total annual carbon flux (A_{tot}) to the biosphere reconstructed for Foulden Maar (Fig. 5c). Early Miocene trees at 50°S likely assimilated $A_{tot} Q_1 - Q_3 =$ 583 265–696 gC m⁻² yr⁻¹, in comparison to $A_{tot} Q_1 - Q_3 = 108 - 182$ gC m⁻² yr⁻¹ in modern evergreen forests, and 584 $A_{tot} Q_1 - Q_3 = 249 - 410 \text{ gC m}^{-2} \text{ yr}^{-1}$ in modern deciduous forests at the same latitude (Fig. 5c). Early 585 Miocene tropical trees appear to have slightly higher total annual carbon flux ($A_{tot} Q_1 - Q_3 = 596 - 1220 \text{ gC}$ 586 $m^{-2} yr^{-1}$) than today (A_{tot} Q₁-Q₃ = 329-721 gC m⁻² yr⁻¹), which, with a year-round growing season in the 587 early Miocene (like today), is likely attributable to a leaf-level fertilization effect, similar to what is 588 589 observed in modern carbon fertilization experiments (Norby et al., 2003; Bader et al., 2013; Yang et al., 590 2016). Although this estimate cannot take the number of leaves per unit area into account, these results suggest enhanced leaf-level productivity during higher than modern C_a in the early Miocene. 591 592 The methods used in this study provide an alternate approach to controlled carbon fertilization 593 experiments, such as the Free Air Carbon Enrichment (FACE) experiments (e.g. Long et al., 2004), toward investigating the effect of increased Ca on the biosphere. FACE experiments provide data on the 594 physiological effects of carbon enrichment on species that evolved under, or had thousands of years to 595 596 adapt to pre-industrial $C_a \approx 280$ ppm), and the physiological changes detected in canopy species are 597 measured as a direct response or over leaf generations (e.g. Norby et al., 2003; Yang et al., 2016). By contrast, our data provide an insight into species that evolved under higher than pre-industrial C_a and had 598

599 many generations of individuals to adapt to incrementally slow changes. The direct, or multi-year

600 physiological response of modern forest trees to enhanced C_a is non-linear and non-uniform (e.g. Long et

al., 2004; Ainsworth and Long, 2005), and therefore further investigations into the physiology of ancient

602 plants operating in high-CO₂ worlds are needed to reveal the complexity of plant responses over

603 evolutionary timescales.

604

605 4 Conclusions

607 than pre-industrial levels at 450–550 ppm, further solidifying the growing consensus of relatively high

608 early Miocene global temperatures maintained by high atmospheric CO₂ (Kürschner et al., 2009;

609 Tesfamichael et al., 2017; Super et al., 2018; Londoño et al., 2018; Moraweck et al., 2019). A relatively

high C_a in the early Miocene also satisfies an Earth System Sensitivity of 3–7°C (Hansen et al., 2013;

611 Royer, 2016). A potential shift in atmospheric CO₂ from 450 to 550, and back to 450, is recorded in the

612 100 kyr of sedimentation and leaf deposition at Foulden Maar. A disruption of the regional carbon and

613 hydrological cycle was also recorded in leaf-wax δ^{13} C and δ D (Reichgelt et al., 2016), and may be linked

to the Antarctic deglaciation at the termination of the Mi-1 (DeConto et al., 2008; Fox et al., 2015;

615 Liebrand et al., 2017).

616 The first record is provided of increased Miocene leaf-level intrinsic water-use efficiency in both temperate New Zealand and the tropics, and we provide evidence for increased leaf-level productivity in 617 618 temperate New Zealand. Enhanced productivity and water-use efficiency on other landmasses in 619 temperate latitudes during the early Miocene, such as North America, Australia, and Asia, would have 620 had a major impact on the global carbon and water cycles. Our gas-exchange results from New Zealand, supplemented with results from Ethiopia (Tesfamichael et al., 2017) and Panama (Londoño et al., 2018) 621 622 provide empirical evidence for high water-use efficiency in the globally warmer world of the early 623 Miocene, associated with elevated C_a. Tropical trees with high water-use efficiency compared to modern, would have likely facilitated forest survival in climates where currently tropical savannas and grasslands 624

exist. These high water-use efficiency forests in the tropics likely persisted until the late Miocene when reduced C_a (Mejía et al., 2017) started favoring the expansion of grasslands, in particular grasslands with the C_4 pathway that is more efficient under low C_a and high temperatures (Strömberg, 2011; Polissar et al., 2016).

Emission scenarios suggest that atmospheric CO_2 will reach our reconstructed early Miocene values of 450 ppm by 2030–2040 CE. While the global temperature response may lag the C_a increase, and forest habitat expansion is hampered by the slow dispersal and growth rate of climax forest trees and anthropogenic influence (e.g., forest fragmentation and fire), early Miocene water-use efficiency and productivity estimates provide insight into future-biosphere potential, as well as into selective pressures that influence the types of plants that may proliferate under future elevated C_a.

635

Acknowledgments. We thank the Gibson family for kindly allowing us access to the site. Funding for
this research was provided by a Royal Society of New Zealand Marsden grant (UOO1115) to DEL, an
NSF grant (EAR13-49659) to WJD, a Vetlesen Foundation Climate Center grant to TR and WJD, and the
Lamont-Doherty Earth Observatory Summer Internship Program for Undergraduates awarded to ACVM.
Wei Huang, Andy Juhl and Nicole DeRoberts are acknowledged for technical support. Gregory Retallack
and an anonymous reviewer are acknowledged for their insightful comments that greatly improved this
manuscript in the review process.

643

Author contributions. TR and WJD conceived of the idea and performed data analyses. BRSF and DEL
collected sediment core, BRSF and TR sampled the sediment core, JGC and JMB identified fossil leaf
taxa. ACVM and TR gathered data from fossil leaves. TR and WJD wrote the paper and all authors
contributed to the final manuscript.

648

649 **Competing interests.** The authors declare no competing interests.

651 References

- Ainsworth, E. A. and Long, S. P.: What have we learned from 15 years of free-air CO₂ enrichment
 (FACE)? A meta-analytic review of the responses of photosynthesis, canopy properties and plant
 production to rising CO₂, New Phytologist, 165, 351–372, 2005.
- Askin, R. A. and Raine, J. I.: Oligocene and early Miocene terrestrial palynology of the Cape Roberts
 drillhole CRP-2/2A, Victoria Land Basin, Antarctica, Terra Antarctica, 7, 493–501, 2000.
- Bader, M. K.-F., Leuzinger, S., Keel, S. G., Siegwolf, R. T. W., Hagedorn, F., Schleppi, P. and Körner,
 C.: Central European hardwood trees in a high CO₂ future: synthesis of an 8-year forest canopy CO₂
 enrichment project, Journal of Ecology, 101, 1509–1519, 2013.
- Bannister, J. M., Conran, J. G., and Lee, D. E.: Lauraceae from rainforest surrounding an early Miocene
 maar lake, Otago, southern New Zealand, Review of Palaeobotany and Palynology, 178, 13–34, 2012.
- Bar-On, Y. M., Philips, R., and Milo, R.: The biomass distribution on Earth, PNAS, 115, 6506–6511,
 2018.
- Beer, C., Reichstein, M., Tomelleri, E., Ciais, P., Jung, M., Carvalhais, N., Rödenbeck, C., Arain, M. A.,
 Baldocchi, D., Bonan, G. B., Bondeau, A., Cescatti, A., Lasslop, G., Lindroth, A., Lomas, M.,
- Luyssaert, S., Margolis, H., Oleson, K. W., Roupsard, O., Veenendaal, E., Viovy, N., Williams, C. J.,
 Woodward, F. I., and Papale, D.: Terrestrial gross ccarbon dioxide uptake: Global distribution and
 covariation with climate, Science, 329, 834–838, 2010.
- Bolton, C. T., Hernández-Sánchez, M. T., Fuertes, M.-Á., González-Lemos, S., Abrevaya, L., MendezVicente, A., Flores, J.-A., Probert, I., Giosan, L., Johnson, J., and Stoll, H. M.: Decrease in
 coccolithophore calcification and CO₂ since the middle Miocene, Nature Communications, 7, 1–13,
 2016.
- Boyden, J. A., Müller, R. D., Gurnis, M., Torsvik, T. H., Clark, J. A., Turner, M., Ivey-Law, H., Watson,
 R. J., and Cannon, J. S.: Next-generation plate-tectonic reconstructions using GPlates. In:
- 675 Geoinformatics: Cyberinfrastructures for the Solid Earth Sciences, Keller, G. R. and Baru, C. (Eds.),
 676 Cambridge University Press, 2011.
- Bush, R. T., Wallace, J., Currano, E. D., Jacobs, B. F., McInerney, F. A., Dunn, R. E., and Tabor, N. J.:
 Cell anatomy and leaf δ¹³C as proxies for shading and canopy structure in a Miocene forest from
 Ethiopia, Palaeogeography, Palaeoclimatology, Palaeoecology, 485, 593–604, 2017.
- 680 Cernusak, L. A.: Gas exchange and water-use efficiency in plant canopies, Plant Biology, 22, 52–67,
 681 2020.
- Conran, J. G., Bannister, J. M., and Lee, D. E.: Fruits and leaves with cuticle of *Laurelia otagoensis* sp.
 nov. (Atherospermataceae) from the early Miocene of Otago (New Zealand), Alcheringa, 37, 1–14,
 2013.
- 685 Conran, J. G., Lee, W. G., Lee, D. E., Bannister, J. M., and Kaulfuss, U.: Reproductive niche
 686 conservatism in the isolated New Zealand flora over 23 million years, Biology Letters, 10, 20140647,
 687 2014.
- Conran, J. G., Bannister, J. M., Mildenhall, D. C., and Lee, D. E.: *Hedycarya* macrofossils and associated
 Planarpollenites pollen from the early Miocene of New Zealand, American Journal of Botany, 103,
 938–956, 2016.
- 691 Cornwell, W. K., Wright, I. J., Turner, J., Maire, V., Barbour, M. M., Cernusak, L. A., Dawson, T.,
- 692 Ellsworth, D., Farquhar, G. D., Griffiths, H., Keitel, C., Knohl, A., Reich, P. B., Williams, D. G.,
- 693 Bhaskar, R., Cornelissen, J. H. C., Richards, A., Schmidt, S., Valladares, F., Körner, C., Schulze, E.-

- D., Buchmann, N., and Santiago, L. S.: Climate and oils together regulate photosynthetic carbon
- isotope discrimination within C₃ plants worldwide, Global Ecology and Biogeography, 27, 1056–
 1067, 2018.
- 697 DeConto, R. M., Pollard, D., Wilson, P. A., Pälike, H., Lear, C. H., and Pagani, M.: Thresholds for
 698 Cenozoic bipolar glaciation, Nature, 455, 652–656, 2008.
- De Lange, P. J.: Fact Sheet (content continuously updated). New Zealand Plant Conservation Network.
 <u>https://www.nzpcn.org.nz</u> (27 May, 2020).
- Dow, G. J., Bergmann, D. C., and Berry, J. A.: An integrated model of stomatal development and leaf
 physiology, New Phytologist, 201, 1218–1226, 2014.
- Drake, B. G., Gonzàlez-Meler, M. A., and Long, S. P.: More efficient plants: A consequence of rising
 atmospheric CO₂?, Annual Review of Plant Physiology and Plant Molecular Biology, 48, 606–639,
 1997.
- Evans, J. R. and Von Caemmerer, S.: Carbon dioxide diffusion inside leaves, Plant Physiology, 110, 339–
 346, 1996.
- Farquhar, G. D., von Caemmerer, S., and Berry, J. A.: A biochemical model of photosynthetic CO₂
 assimilation in leaves of C₃ species, Planta, 149, 78–90, 1980.
- Farquhar, G. D., O'Leary, M. H., and Berry, J. A.: On the relationship between carbon isotope
 discrimination and the intercellular carbon dioxide concentration in leaves, Australian Journal of Plant
 Physiology, 9, 121–137, 1982.
- Farquhar, G. D. and Richards, R. A.: Isotopic composition of plant carbon correlates with water-use
 efficiency of wheat genotypes, Australian Journal of Plant Physiology, 11, 539–552, 1984.
- Farquhar, G. D., Ehleringer, J. R., and Hubick, K. T.: Carbon isotope discrimination and photosynthesis,
 Annual Review of Plant Physiology and Plant Molecular Biology, 40, 503–537, 1989.
- Foster, G. L., Royer, D. L., and Lunt, D. J.: Future climate focing potentially without precedent in the last
 420 million years, Nature Communications, 8, 14845, 2017.
- Feng, X.: Trends in intrinsic water-use efficiency of natural trees for the past 100–200 years: a response
 to atmospheric CO₂ concentration, Geochimica et Cosmochimica Acta, 63, 1891–1903, 1999.
- Fox, B. R. S., Wartho, J.-A., Wilson, G. S., Lee, D. E., Nelson, F. E., and Kaulfuss, U.: Long-term
 evolution of an Oligocene/Miocene maar lake from Otago, New Zealand, Geochemistry, Geophysics,
 Geosystems, 16, 59–76, 2015.
- Fox, B. R. S., Wilson, G. S., and Lee, D. E.: A unique annually laminated maar lake sediment record
 shows orbital control of Southern Hemisphere midlatitude climate across the Oligocene-Miocene
 boundary, Geological Society of America Bulletin, 128, 609–626, 2016.
- 727 Frank, D. C., Poulter, B., Saurer, M., Esper, J., Huntingford, C., Helle, G., Treydte, K., Zimmermann, N.
- E., Schleser, G. H., Ahlström, A., Ciais, P., Friedlingstein, P., Levis, S., Lomas, M., Sitch, S., Viovy,
- N., Andreu-Hayles, L., Bednarz, Z., Berninger, F., Boettger, T., D'Alessandro, H., Kalela-Brundin, M.,
 Krapiec, M., Leuenberger, M., Loader, N. J., Marah, H., Masson-Delmotte, V., Pazdur, A., Pawelczyk,
- Krapiec, M., Leuenberger, M., Loader, N. J., Marah, H., Masson-Delmotte, V., Pazdur, A., Pawelczyk,
 S., Pierre, M., Planells, O., Pukiene, R., Reynolds-Henne, C. E., Rinne, K. T., Saracino, A., Sonninen,
- Figure 1. Stievenard, M., Switsur, V. R., Szczepanek, E., Szychowska-Krapiec, L., Todaro, L., Waterhouse,
- J. S., and Weigl, M.: Water-use efficiency and transpiration across European forests during the
 Anthropocene, Nature Climate Change, 5, 579–583, 2015.
- Franks, P. J. and Beerling, D. J.: Maximum leaf conductance driven by CO₂ effects on stomatal size and density over geologic time. DNAS, 106, 10242, 10247, 2000
- 736
 density over geologic time, PNAS, 106, 10343–10347, 2009.

- 737 Franks, P. J., Drake, P. L., and Beerling, D. J.: Plasticity in maximum stomatal conductance constrained
- by negative correlation between stomatal size and density: an analysis using *Eucalyptus globulus*,
 Plant, Cell and Environment, 32, 1737–1748, 2009.
- 740 Franks, P. J., Adams, M. A., Amthor, J. S., Barbour, M. M., Berry, J. A., Ellsworth, D. S., Farquhar, G.
- D., Ghannoum, O., Lloyd, J., McDowell, N. G., Norby, R. J., Tissue, D. T., and von Caemmerer, S.:
 Sensitivity of plants to changing atmospheric CO₂ concentration: from the geological past to the next
 century, New Phytologist, 197, 1077–1094, 2013.
- Franks, P. J., Royer, D. L., Beerling, D. J., van de Water, P. K., Cantrill, D. J., Barbour, M. M., and
 Berry, J. A.: New constraints on atmospheric CO₂ concentration for the Phanerozoic, Geophysical
 Research Letters, 41, 4685–4694, 2014.
- Gimeno, T. E., McVicar, T. R., O'Grady, A. P., Tissue, D. T. and Ellsworth, D. S.: Elevated CO₂ did not affect the hydrological balance of a mature native *Eucalyptus* woodland, Global Change Biology, 24, 3010–3024, 2018.
- Graham, H. V., Patzkowsky, M. E., Wing, S. L., Parker, G. G., Fogel, M. L., and Freeman, K. H.:
 Isotopic characteristics of canopies in simulated leaf assemblages, Geochimica et Cosmochimica Acta,
 144, 82–95, 2014.
- Greenop, R., Sosdian, S. M., Henehan, M. J., Wilson, P. A., Lear, C. H., and Foster, G. L.: Orbital
 forcing, ice volume, and CO₂ across the Oligocene–Miocene transition, Paleoceanography and
 Paleoclimatology, 34, 316–628, 2019.
- Hansen, J., Sato, M., Russell, G., and Kharecha, P.: Climate sensitivity, sea level and atmospheric carbon
 dioxide, Philosophical Transactions of the Royal Society A, 371, 20120294, 2013.
- Henrot, A.-J., François, L., Favre, E., Butzin, M., Ouberdous, M., and Munhoven, G.: Effects of CO₂,
 continental distribution, topography and vegetation changes on the climate at the Middle Miocene: a
 model study, Climate of the Past, 6, 675–694, 2010.
- Herold, N., Müller, R. D., and Seton, M.: Comparing early to middle Miocene terrestrial climate
 simulations with geological data, Geosphere, 6, 952–961, 2010.
- Ji, S., Nie, J., Lechler, A., Huntington, K. W., Heitmann, E. O., and Breecker, D. O.: A symmetrical CO₂
 peak and asymmetrical climate change during the middle Miocene, Earth and Planetary Science
 Letters, 499, 134–144, 2018.
- Kaulfuss, U.: Crater stratigraphy and the post-eruptive evolution of Foulden Maar, southern New
 Zealand, New Zealand Journal of Geology and Geophysics, 60, 410–432, 2017.
- Kaulfuss, U., Lee, D. E., Barratt, B. I. P., Leschen, R. A. B., Larivière, M.-C., Dlussky, G. M.,
 Henderson, I. M., and Harris, A. C.: A diverse fossil terrestrial arthropod fauna from New Zealand:
 evidence from the early Miocene Foulden Maar fossil lagerstätte, Lethaia, 48, 299–308, 2015.
- Kohn, M.J.: Carbon isotope discrimination in C3 land plants is independent of natural variations in p_{CO2},
 Geophysical Perspective Letters, 2, 35–43. 2016.
- Kürschner, W. M.: The anatomical diversity of recent and fossil leaves of the durmast oak (*Quercus petraea* Lieblein/*Q. pseudocastanaea* Goeppert) implications for their use as biosensors of
 palaeoatmospheric CO₂ levels, Review of Palaeobotany and Palynology, 96, 1–30, 1997.
- Kürschner, W. M., Kvaček, Z., and Dilcher, D. L.: The impact of Miocene atmospheric carbon dioxide
 fluctuations on climate and the evolution of terrestrial ecosystems, PNAS, 105, 449–453, 2008.
- 778 Kürschner, W. M. and Kvaček, Z.: Oligocene-Miocene CO₂ fluctuations, climatic and palaeofloristic
- trends inferred from fossil plant assemblages in central Europe, Bulletin of Geosciences, 84, 189–202,
 2009.

- Leathwick, J. R.: New Zealand's potential forest pattern as predicted from current species-environment
 relationships, New Zealand Journal of Botany, 39, 447–464, 2001.
- Lee, D. E., Lindqvist, J. K., Beu, A. G., Robinson, J. H., Ayress, M. A., Morgans, H. E. G., and Stein, J.
 K.: Geological setting and diverse fauna of a Late Oligocene rocky shore ecosystem, Cosy Dell,
 Southland, New Zealand Journal of Geology and Geophysics, 57, 195–208, 2014.
- Lee, D. E., Kaulfuss, U., Conran, J. G., Bannister, J. M., and Lindqvist, J. K.: Biodiversity and
 palaeoecology of Foulden Maar: an early Miocene *Konservat-Lagerstätte* deposit in southern New
 Zealand, Alcheringa, 40, 525–541, 2016.
- Liebrand, D., de Bakker, A. T. M., Beddow, H. M., Wilson, P. A., Bohaty, S. M., Ruessink, G., Pälike,
 H., Batenburg, S. J., Hilgen, F. J., Hodell, D. A., Huck, C. E., Kroon, D., Raffi, I., Saes, M. J. M., van
 Dijk, A. E., and Lourens, L. J.: Evolution of the early Antarctic ice ages, PNAS, 114, 3867–3872,
 2017.
- Lindqvist, J. K. and Lee, D. E.: High-frequency paleoclimate signals from Foulden Maar, Waipiata
 Volcanic Field, southern New Zealand: An Early Miocene varved lacustrine diatomite deposit,
 Sedimentary Geology, 222, 98–110, 2009.
- Londoño, L., Royer, D. L., Jaramillo, C. A., Escobar, J., Foster, D. A., Cárdenas-Rozo, A. L., and Wood,
 A.: Early Miocene CO₂ estimates from a Neotropical fossil leaf assemblage exceed 400 ppm,
 American Journal of Botany, 105, 1929–1937, 2018.
- Long, S. P., Ainsworth, E. A., Rogers, A. and Ort, D. R.: Rising atmospheric carbon dioxide: Plants
 FACE the future, Annual Review of Plant Biology, 55, 591–628, 2004.
- Maire, V., Wright, I. J., Prentice, I. C., Batjes, N. H., Bhaskar, R., van Bodegom, P. M., Cornwell, W. K.,
 Ellsworth, D. S., Niinemets, Ü., Ordonez, A., Reich, P. B., and Santiago, L. S.: Global effects of soil
 and climate on leaf photosynthetic traits and rates, Global Ecology and Biogeography, 24, 706–717,
 2015.
- Marrero, T. R. and Mason, E. A.: Gaseous diffusion coefficients, Journal of Physical and Chemical
 Reference Data, 1, 3–118, 1972.
- McElwain, J. C., Montañez, I. P., White, J. D., Wilson, J. P., and Yiotis, C.: Was atmospheric CO₂ capped
 at 1000 ppm over the past 300 million years?, Palaeogeography, Palaeoclimatology, Palaeoecology,
 441, 653–658, 2016.
- Mejía, L. M., Méndez-Vicente, A., Abrevaya, L., Lawrence, K. T., Ladlow, C., Bolton, C., Cacho, I. and
 Stoll, H.: A diatom record of CO₂ decline since the late Miocene, Earth and Planetary Science Letters,
 479, 18–33, 2017.
- Moraweck, K., Grein, M., Konrad, W., Kvaček, J., Kovar-Eder, J., Neinhuis, C., Traiser, C., and
 Kunzmann, L.: Leaf traits of long-ranging Paleogeone species and their relationship with depositional
- facies, climate an atmospheric CO_2 level, Palaeontographica Abteilung B, 298, 93–172, 2019.
- 816 Németh, K. and White, J. D. L.: Reconstructing eruption processes of a Miocene monogenetic volcanic
- field from vent remnants: Waipiata Volcanic Field, South Island, New Zealand, Journal of
 Volcanology and Geothermal Research, 124, 1–21, 2003.
- Niinemets, Ü., Díaz-Espejo, A., Flexas, J., Galmés, J., and Warren, C. R.: Role of mesophyll diffusion
 conductance in constraining potential photosynthetic productivity in the field, Journal of Experimental
 Botany, 60, 2249–2270, 2009.
- 822 Niinemets, Ü., Flexas, J., and Peñuelas, J.: Evergreens favored by higher responsiveness to increased
- 823 CO₂, TRENDS in Ecology and Evolution, 26, 136–142, 2011.

- Norby, R. J., Sholtis, J. D., Gunderson, C. A. and Jawdy, S. S.: Leaf dynamics to a deciduous forest
 canopy: no response to elevated CO₂, Oecologia, 136, 574–584, 2003.
- Polissar, P. J., Rose, C., Uno, K. T., Phelps, S. R., and DeMenocal, P.: Synchronous rise of African C₄
 ecosystems 10 million years ago in the absence of aridification, Nature Geoscience, 12, 657–660,
 2016.
- Reichgelt, T., Kennedy, E. M., Mildenhall, D. C., Conran, J. G., Greenwood, D. R., and Lee, D. E.:
 Quantitative palaeoclimate estimates for Early Miocene southern New Zealand: Evidence from
 Foulden Maar, Palaeogeography, Palaeoclimatology, Palaeoecology, 378, 36–44, 2013.
- Reichgelt, T., D'Andrea, W. J., and Fox, B. R. S.: Abrupt plant physiological changes in southern New
 Zealand at the termination of the Mi-1 event reflect shifts in hydroclimate and *p*CO₂, Earth and
- 834 Planetary Science Letters, 455, 115–124, 2016.
- Reichgelt, T. and D'Andrea, W. J.: Plant carbon assimilation rates in atmospheric CO₂ reconstructions,
 New Phytologist, 223, 1844–1855, 2019.
- Reichgelt, T., Kennedy, E. M., Conran, J. G., Lee, W. G., and Lee, D. E.: The presence of moisture
 deficits in Miocene New Zealand, Global and Planetary Change, 172, 268–277, 2019.
- Retallack, G. J. and Conde, G. D.: Deep time perspective on rising atmospheric CO₂, Global and
 Planetary Change, 189, 103177, 2020.
- Roeske, C. A. and O'Leary, M. H.: Carbon isotope effects on the enzyme-catalyzed carboxylation of
 ribulose bisphosphate, Biochemistry, 23, 6275–6284, 1984.
- Royer, D. L.: Climate sensitivity in the geologic past, Annual Review of Earth and Planetary Sciences,
 44, 277–293, 2016.
- Royer, D. L., Moynihan, K. M., McKee, M. L., Londoño, L., and Franks, P. J.: Sensitivity of a leaf gasexchange model for estimating paleoatmospheric CO₂ concentration, Climate of the Past, 15, 795–809,
 2019.
- Rubel, F. and Kottek, M.: Observed and projected climate shifts 1901–2100 depicted by world maps of
 the Köppen-Geiger climate classification, Meteorologische Zeitschrift, 19, 135–141, 2010.
- Šantrůček, J., Vráblová, M., Šimková, M., Hronková, M., Drtinová, M., Květoň, J., Vrábl, D., Kubásek,
 J., Macková, J., Wiesnerová, D., Neuwithová, J., and Schreiber, L.: Stomatal and pavement cell
- density linked to leaf internal CO₂ concentration, Annals of Botany, 114, 191–202, 2014.
- Schneider, C. A., Rasband, W. S., and Eliceiri, K. W.: NIH Image to ImageJ: 25 years of image analysis,
 Nature Methods, 9, 671–675, 2012.
- Schuepp, P. H.: Transley Review No. 59 Leaf boundary layers, New Phytologist, 125, 477–507, 1993.
- 856 Soh, W. K., Wright, I. J., Bacon, K. L., Lenz, T. I., Steinthorsdottir, M., Parnell, A. C., and McElwain, J.
- 857 C.: Palaeo leaf economics reveal a shift in ecosystem function associated with the end-Triassic mass858 extinction event, Nature Plants, 3, 8, 2017.
- Soh, W. K., Yiotis, C., Murray, M., Parnell, A. C., Wright, I. J., Spicer, R. A., Lawson, T., Caballero, R.,
 and McElwain, J. C.: Rising CO₂ drives divergence in water use efficiency of evergreen and deciduous
 plants, Science Advances, 5, eaax7906, 2019.
- Spicer, R. A., Valdes, P. J., Spicer, T. E. V., Craggs, H. J., Srivastrava, G., Mehrotra, R. C., and Yang, J.:
 New developments in CLAMP: Calibration using global gridded meteorological data,
- Palaeogeography, Palaeoclimatology, Palaeoecology, 283, 91–98, 2009.
- 865 Steinthorsdottir, M., Vajda, V., and Pole, M. S.: Significant transient *p*CO₂ perturbation at the New
- 266 Zealand Oligocene–Miocene transition recorded by fossil plant stomata, Palaeogeography,
- Palaeoclimatology, Palaeoecology, 515, 152–161, 2019.

- Strömberg, C. A. E.: Evolution of grasses and grassland ecosystems, Annual Review of Earth and
 Planetary Sciences, 39, 517–544, 2011.
- Super, J. R., Thomas, E., Pagani, M., Huber, M., O'Brien, C., and Hull, P. M.: North Atlantic temperature
 and *p*CO₂ coupling in the early-middle Miocene, Geology, 46, 519–522, 2018.
- 872 Tesfamichael, T., Jacobs, B. F., Tabor, N. J., Michel, L., Currano, E. D., Feseha, M., Barclay, R. S.,
- Kappelman, J., and Schmitz, M.: Settling the issue of "decoupling" between atmospheric carbon
 dioxide and global temperature: [CO₂]_{atm} reconstructions across the warming Paleogene-Neogene
- divide, Geology, 45, 999–1002, 2017.
- Tipple, B. J., Meyers, S. R., and Pagani, M.: Carbon isotope of Cenozoic CO₂: A comparative evaluation
 of available geochemical proxies, Paleoceanography, 25, PA3202, 2010.
- Witkowski, C. R., Agostini, S., Harvey, B. P., van der Meer, M. T. J., Sinninghe Damsté, J. S., and
 Schouten, S.: Validation of carbon isotope fractionation in algal lipids as a *p*CO₂ proxy using a natural
 CO₂ seep (Shikine Island, Japan), Biogeosciences, 16, 4451–4461, 2019.
- Yang, Y., Donohue, R. J., McVicar, T. R., Roderick, M. L. and Beck, H. E.: Long-term CO₂ fertilization
 increases vegetation productivity and has little effect on hydrological partitioning in tropical
 rainforests, Journal of Geophysical Research: Biogeosciences, 121, 2125–2140, 2016.
- 884 Zachos, J. C., Pagani, M., Sloan, L., Thomas, E., and Billups, K.: Trends, rhythms, and aberrations in
- global climate 65 Ma to present, Science, 292, 686–693, 2001.
- Zhou, S., Yu, B., Shwalm, C. R., Ciais, P., Zhang, Y., Fisher, J. B., Michalak, A. M., Wang, W., Poulter,
 B., Huntzinger, D. N., Niu, S., Mao, J., Jain, A., Ricciuto, D. M., Shi, X., Ito, A., Wei, Y., Huang, Y.,
 and Wang, G.: Response of water use efficiency to global environmental change based on output from
 terrestrial biosphere models, Global Biogeochemical Cycles, 31, 1639–1655, 2017.
- Zhu, Z., Piao, S., Myneni, R. B., Zeng, Z., Canadell, J. G., Ciais, P., Sitch, S., Friedlingstein, P., Arneth,
- 891 A., Cao, C., Cheng, L., Kato, E., Koven, C., Li, Y., Lian, X., Liu, Y., Liu, R., Mao, J., Pan, Y., Peng,
- 892 S., Peñuelas, J., Poulter, B., Pugh, T. A. M., Stocker, B. D., Viovy, N., Wang, X., Wang, Y., Xiao, Z.,
- Yang, H., Zaehle, S., and Zeng, N.: Greening of the Earth and its drivers, Nature Climate Change, 6,
 791–795, 2016.
- 895





Figure 1. Foulden Maar site information. (a) Location of the Foulden Maar deposit and paleogeographic
reconstruction of early Miocene New Zealand (Boyden et al., 2011; Lee et al., 2014). (b) Schematic
reconstruction of the Foulden Maar depositional environment. (c) Stratigraphic column of the Foulden
Maar core (Fox et al., 2015), with sample locations and bulk organic δ¹³C (Reichgelt et al., 2016).

904

905 Fig. 2







913 Myrtaceae (Myrt), and an understory comprising cf. Myrsine (Myrs), cf. Ripogonum (Rip), cf. Dysoxylum

- 914 (Dys), and leaf type "O". Hedycarya pluvisilva (Hplu), Laurelia otagoensis (Lota), Beilschmiedia
- 915 otagoensis (Bota), cf. Cryptocarya (cfC), cf. Endiandra (cfE), and leaf types "C", "H", "P", and "Q"
- 916 could not be ecologically placed with certainty. (c) Probability density distributions of C_a reconstructions
- 917 from canopy (thick light green line) and understory components (thick dark green line) using a gas-
- exchange model (Franks et al., 2014). Grey curves represents the probability distribution of 10,000
- 919 Monte Carlo reconstructions on a single fossil leaf.
- 920





- al., 2014), divided by bulk carbon isotope phases (Fig. 1c).
- 924
- 925 Fig. 4





Figure 4. Neogene Earth System Sensitivity (ESS) and Ca reconstructions. Calculated Ca levels for an 927 ESS range of 3–7 °C (orange shaded area) for the last 30 Ma (a), and for the interval between 22.5–23.5 928 929 Ma (b), the red dashed line in (b) indicates the global average surface temperature (T_s) in the earliest 930 Miocene (Hansen et al., 2013). The ESS envelope was determined using deep-sea δ^{18} O of benthic foraminifera (Zachos et al., 2001) and the transform function approach from Hansen et al. (2013) 931 932 (Supplementary Information). Proxy-based Neogene C_a reconstructions are derived from a previously published compilation (Foster et al., 2017) and are supplemented with more recently published data (Ji 933 et al., 2019; Londoño et al., 2018; Super et al., 2018; Greenop et al., 2019; Moraweck et al., 2019, 934 935 Steinthorsdottir et al., 2019). Error bars on gas-exchange based proxy estimates represent $\pm 1\sigma$. 936

937 Fig. 5



939 Figure 5. Early Miocene leaf-level physiological parameters of canopy trees. (a) Intrinsic water-use 940 efficiency (iWUE) of evergreen (green circles) and deciduous trees (red circles) based on modern leaf-941 level measurements (Maire et al., 2015), and fossil reconstructions (green triangles and diamonds). Error bars on fossil-derived data indicate $\pm 1\sigma$, box-and-whisker plots indicate median, first and third quartile 942 (O1 and O3), and 95% confidence interval of modern leaves of canopy trees. Individual datapoints are 943 944 randomized on the x-axis for a clearer depiction of the distribution. (b) Conductance to water (G_w) from 945 modern evergreen (Ev) and deciduous (Dec) trees (Maire et al., 2015) and fossils (EM) from different latitudes. The shaded red and green areas indicate the Q1–Q3 range of modern evergreen and deciduous 946 trees, respectively, and the dashed lines indicate the overall linear trend with latitude. Text in panel is the 947 O1–O3 range for each group, grouped in 5° latitude bins. (c) Total annual carbon flux per unit leaf area 948 949 (Atot) from modern evergreen (Ev) and deciduous (Dec) trees (Maire et al., 2015) and fossils (EM) from different latitudes. The shaded red and green areas indicate the Q1-Q3 range of modern evergreen and 950 951 deciduous trees, respectively, and the dashed lines indicate the overall exponential trend with latitude. 952 Text in panel is the Q1–Q3 range for each group, grouped in 5° latitude bins.

Data availability. All raw measurement data on fossil leaves generated for this paper is available in the online supplementary information. Raw measurements on fossil leaves from Ethiopia (Tesfamichael et al., 2017) and Panama (Londoño et al., 2018), δ^{18} O measurements (Zachos et al., 2001), and iWUE, G_w

- and A_n measurements on modern plants (Maire et al., 2015) are available through the cited original
- 958 works.